Transparent Conducting Oxides

David S. Ginley and Clark Bright, Guest Editors

In the interim between the conception of this issue of *MRS Bulletin* on transparent conducting oxides (TCOs) and its publication, the remarkable applications dependent on these materials have continued to make sweeping strides. These include the advent of larger flat-screen high-definition televisions (HDTVs), larger and higher-resolution screens on portable computers, the increasing importance of low emissivity ("low-e") and electrochromic windows, a significant increase in the manufacturing of thin-film photovoltaics (PV), and a plethora of new hand-held and smart devices, all with smart displays.[1-7] Coupled with the increased importance of TCO materials to these application technologies has been a renaissance over the last two years in the science of these materials. This has included new *n*-type materials, the synthesis of true *p*-type materials, and the theoretical prediction and subsequent confirmation of the applicability of codoping to produce *p*-type ZnO. Considering that over the last 20 years much of the work on TCOs was empirical and focused on ZnO and variants of $In_xSn_{1-x}O_y$, it is quite remarkable how this field has exploded. This may be a function of not only the need to achieve higher performance levels for these devices, but also of the increasing importance of transition-metal-based oxides in electro-optical devices. This issue of *MRS Bulletin* is thus well timed to provide an overview of this rapidly expanding area. Included are articles that cover the industrial perspective, new *n*-type materials, new *p*-type materials, novel deposition methods, and approaches to developing both an improved basic understanding of the materials themselves as well as models capable of predicting performance limits.

The current TCO industry is dominated by just a few materials. We will present an overview of the current state of the field, in order to help the reader develop an appreciation for the size and demands of the industry as well as the need for new materials.

The two dominant markets for TCOs are in architectural applications and flat-panel displays (FPDs). The architectural use of TCOs is for energy-efficient windows. Fluorine-doped tin oxide, deposited by a pyrolytic process, is the TCO most often used in this application. Windows with tin oxide coatings are efficient in preventing radiative heat loss, due to tin oxide's low emissivity of about 0.16. Such "low-e" windows are ideal for cold or moderate climates. In addition, pyrolytic tin oxide is used for coating heated glass freezer doors in commercial use. The annual consumption (in 1996) of TCO-coated glass (primarily for low-e coatings) in the United States was 7.3×10^7 m^2, or >27 mi^2.[8] Added to this output are the increasing amounts used in displays and PVs.

Pyrolytic tin oxide is also used in PV modules, touch screens, and plasma displays. However, indium tin oxide (ITO) is the TCO used most often in the majority of FPD applications. In FPDs, the basic function of ITO is as a transparent electrode. Often, the ITO will have additional functions, for example, as an antistatic electromagnetic interference shield or an electric heater.

The volume of FPDs produced, and hence the volume of TCO (ITO) coatings produced, continues to grow rapidly. The market for FPDs in 2000 is estimated to be over $15 billion and is predicted to grow to over $27 billion by 2005. The forecast for growth in market size is shown in Figure 1.

Large quantities of TCOs also are deposited onto plastic film in vacuum roll-to-roll coating processes. Again, the major TCO material produced for a variety of these applications is ITO. Industrial issues for current applications and their requirements will be summarized in this issue by Brian Lewis from Arconium Alpha-Fry Technologies and David Paine from Brown University.

In the last few years, the perception that ZnO- and InSnO-based materials were sufficient for TCO applications has begun to change. As is the case in many technological areas, this is a consequence of the acknowledgment of the limitations of the existing materials as well as a realization that new materials can open the way to new and improved devices. Coupled with this has been an improved capability, stimulated in part by the development of high-temperature superconducting materials, for the synthesis, thin-film deposition, and characterization of oxide-based materials.

Limitations of the existing materials become more critical in view of the increasing need for larger-area display devices with greater writing speeds. As the screen size of flat-panel televisions increases, and faster graphics are required on portable computers, it becomes increasingly important to decrease resistivity while maintaining transparency in the TCO layers.[1,2,9] Figure 2 illustrates a simple TCO transmission curve, showing both the band-edge and plasma-edge limits for two SnO_2 films with different resistivities. Clearly, as the resistivity decreases, the plasma edge moves to higher energy, decreasing transmission in the infrared.[10]

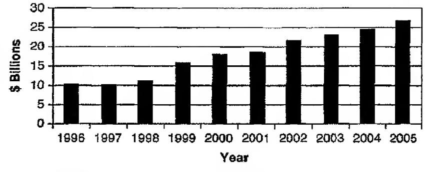

Figure 1. Market forecast for flat-panel display sales (data from DisplaySearch).

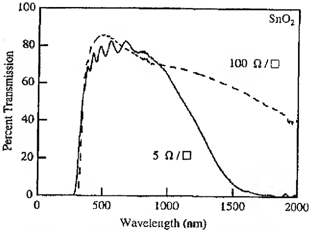

Figure 2. Optical transparency versus conductivity for two SnO_2 films. As the resistivity decreases, long-wavelength transparency also decreases.

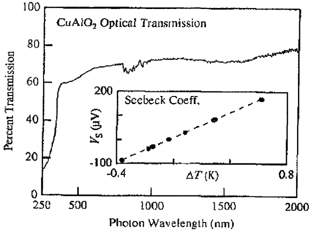

Figure 3. Optical transparency and Seebeck data for a 300–500-nm-thick film of p-type $CuAlO_2$.

Accomplishing the goal of increased conductivity and transparency will require a deeper understanding of the relationships between the structure and the electro-optical properties of these materials and will probably require new materials as well.

The demand for new materials is also amplified by a variety of potential new uses for TCOs; these include novel applications in more demanding environments than those currently and new heterostructure applications as part of the rapid emergence of all-oxide electronics. Thus TCOs may be in demand not just for their electro-optical properties, but for their interfacial and material-compatibility properties as well.

An example is the use of TCOs in a superstrate CdTe solar cell. Since the TCO is deposited as one of the first layers of a PV cell, followed by the CdS and CdTe layers, the TCO must survive the demanding processing environment required for the rest of the cell. Recent results have shown that the use of more stable Cd_2SnO_4 can result in significant improvements in device efficiencies.[11,12] Similarly, considerable interest exists in developing *p*-type TCOs. These would open the way not only to a new generation of transparent electrical contacts for *p*-type semiconductors such as amorphous Si, but also, in combination with *n*-type materials, to transparent-oxide electronics.

Until very recently, little work had been done on the development of *p*-type TCOs, but over the last two years, a number of significant developments have come about. The group of Kawazoe et al. has published papers on $CuAlO_2$ and Cu_2SrO_2 as true *p*-type TCOs.[13,14] The optical characteristics and Seebeck measurements of a thin $CuAlO_2$ film are illustrated in Figure 3.[15] The positive values of the Seebeck coefficient indicate that the majority of carriers are, indeed, holes. Interestingly, it appears that, similar to the copper-based high-T_c superconductors (HTSs), the existence of CuO planes is critical for many of the electronic properties. These new materials offer the potential for a variety of new devices. $CuAlO_2$, although hard to make, is a very stable material, and the Sr-based material can be processed at temperatures as low as 200°C.[14–16] Also, it may soon be possible to dope existing *n*-type TCOs to produce *p*-type materials. Initial results on ZnO showed that small amounts of nitrogen could be incorporated to form a *p*-type semiconductor. Theoretical results for the III-V and II-VI materials subsequently indicated that codoping of these materials may allow not only for type conversion, but also high doping levels. A recent report from Japan confirms this: by codoping ZnO with N and Ga,[17,18] the authors obtained carrier concentrations as high as 10^{19} cm^{-3}.[19] The combination of these new materials and new approaches to *p*-type doping of oxides leads to the hope of adding highly conductive *p*-type TCOs to the device designers' toolbox. The *p*-type TCO area will be reviewed in the article by Hiroshi Kawazoe, Hiroshi Yanagi, Kasushige Ueda, and Hideo Hosono from Hoya Corporation and the Tokyo Institute of Technology.

The primary *n*-type TCOs have remained virtually unchanged for 20 years. The principal constituents have been simple or binary oxides of SnO_2, In_2O_3, ZnO, and CdO. Tin oxide, ITO, and F- or Al-doped ZnO have been the principal commercial TCOs.[1,2] As synthesis techniques for oxides have advanced, owing to the large push in HTS materials, other oxide-related areas such as ferroelectrics, dielectric materials, and TCOs have benefited. Recent work has begun to explore new binary-oxide combinations and even to move into phase fields of ternaries. Cd_2SnO_4, Zn_2SnO_4, $MgIn_2O_4$, $CdSb_2O_6$:Y, $ZnSnO_3$, $GaInO_3$, $Zn_2In_2O_5$, and $In_4Sn_3O_{12}$ are just a few of the ternary *n*-type materials under investigation.[12,20–25] Many of these materials have shown some improved properties over the established materials, although none of them has yet had sufficient overall properties to clearly replace the existing commercial materials. Figure 4 illustrates a comparison between conventional SnO_2 films (carrier density $n = 5 \times 10^{20}$ cm^{-3}; mobility $\mu = 15$ cm^2 V^{-1} S^{-1}; resistance $R = 16.7\ \Omega/\square$) and Cd_2SnO_4 films ($n = 3.2 \times 10^{20}$ cm^{-3}, $\mu = 54$ cm^2 V^{-1} S^{-1}, and $R = 7.2\ \Omega/\square$).[11]

Cd_2SnO_4, despite having a somewhat lower carrier concentration than SnO_2, is actually a better conductor due to its higher carrier mobility. This results in overall improved performance. Cd_2SnO_4 is also a more robust material and can survive in process environments where SnO_2-based materials have difficulties. It is, in fact, more readily etchable and generally smoother than SnO_2 films. This particular example illustrates the complex nature of the electro-optical properties in these materials and their potential for improvement.

In addition to the exploration of new materials, several researchers have investigated new dopants for existing materials. For example, ZnO:Ga films have shown higher performance levels than previously obtained, with resistivities of about 10^{-4} Ω cm. Furthermore, many are realizing that moving into ternary systems may make it possible to design new materials with improved properties for a particular application. The existing areas for new *n*-type materials are discussed in the articles by Tadatsugu Minami of the Kanazawa Institute of Technology and by Arthur J. Freeman, Kenneth R. Poeppelmeier, Thomas O. Mason, Robert P.H. Chang, and Tobin J. Marks of Northwestern University.

One area of particular interest, given these new materials, is how to effectively deposit them, especially for large-area applications. Conventional SnO_2 TCOs often use liquid precursors sprayed on hot glass. ZnO-based materials are often sputtered. The utility of the new materials may depend on the development of improved approaches to the deposition of thin films. This can include new methods, such as pulsed laser deposition, and the development of new sources for conventional chemical vapor deposition (CVD)-type methods.[4,26–28] For CVD, considerable novel chemistry is required to balance the demand for source materials with sufficient volatility and reactivity to provide high rates of deposition and yield pure materials. Some recent work in this area is summarized in the article by Freeman et al.

A pivotal requirement for the continued and expanded application of transparent conductors is an increased understanding of the fundamental interrelationships between conductivity and transparency, and from this, the development of models of the performance limits of these materials. Ideally, a set of modeling tools will be developed to help not only in designing devices incorporating TCOs, but also in developing optimized materials. With the exception of a few stalwarts working in this area, the basic science behind TCOs has not received much emphasis. Due to the success of new materials and the increased demands on TCO performance, this is starting to change. The TCO science area has also benefited from the array of tools developed to characterize oxides in the high-T_c area. We present two articles in this field. One addresses the issue of the performance limits of TCOs.[4,9,26–31] Roy G. Gordon from Harvard University addresses the complex issue of balancing the material and electro-optical properties of TCOs for various applications. For each application, the most suitable TCO is one that best meets particular criteria, including optical, electrical, mechanical, chemical, and/or economic factors. The differences among these criteria have led to different choices of TCOs for different applications. Fundamental physical constraints on TCOs limit the possibilities of satisfying these criteria more closely. Through understanding these relationships, we can assess the ability of the current materials to meet applications needs and the ability of next-generation materials to supplant them.

Coupled with this development of descriptive property models for TCOs is the necessity of understanding the nature of doping and conductivity in these materials. It is crucial to better understand the origin of the carriers in the TCOs and the dominant scattering mechanisms limiting performance, especially in the critical area of the mobility of the carriers.[32] In order to optimize performance in most applications, it is necessary to determine the optimum combination of mobility and carrier concentration, thus achieving the necessary conductivity and transparency. However, this must be done without introducing midgap states that might affect transparency. This can be a difficult challenge in materials that are deposited at low temperatures and have very fine grain structures or, in some cases, are even amorphous. To develop an understanding of the complex relationships between scattering and the structural properties of TCOs requires the application of more in-depth characterization than has been applied previously. In some cases, the development of new analytical approaches is required. Measurement techniques include optical and Fourier transform infrared spectrometries, spectroscopic ellipsometry, x-ray diffraction, high-resolution electron microscopy, Mössbauer and Raman spectroscopies, and a novel thermogalvanomagnetic characterization technique.[32] These collective measurements are providing a better un-

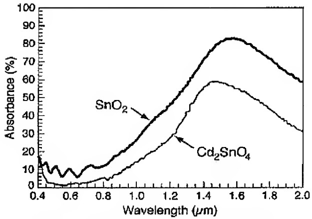

Figure 4. The absorbance of Cd_2SnO_4 compared with that of typical SnO_2 films. The lower carrier concentration in Cd_2SnO_4 results in increased transparency.

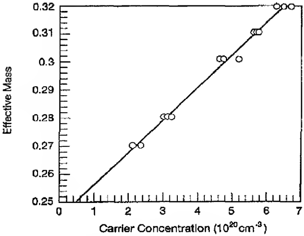

Figure 5. Effective mass of electrons in Cd_2SnO_4 films as a function of their carrier concentration. This indicates that the conduction band of the Cd_2SnO_4 is nonparabolic.

derstanding of the source of carriers and the dominant scattering mechanisms in TCOs. This is illustrated in Figure 5 for Cd_2SnO_4, with data taken by the method of four coefficients. This area is reviewed in the article by Timothy J. Coutts, David L. Young, and Xiaonan Li from the National Renewable Energy Laboratory.

This improved understanding should allow the fabrication of all-oxide devices with the potential advantages of transparency, high-temperature performance, and radiation hardness.[33] Finally, although it is clear that the near future of transparent conductive coatings is based on continued development of oxide-based materials, the next generation of transparent conductors may be based on completely different materials. Researchers are beginning to investigate organic materials and wide-gap, nonoxide materials and to rediscover thin-film metallic layers.[34–43] As the articles in this issue illustrate, the horizon for improved TCOs and oxide electronics is much closer than it was just two years ago.

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